PHOTOCHEMICAL POLLUTION MODELLING IN COMPLEX TERRAIN

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INTRODUCTION

Ozone is considered one of the most significant pollutants with respect to the potential impacts to human health and natural ecosystem, both in terms of critical episodes and as long-term exposures. Consequently, in order to assess the comprehensive effects of photochemical pollution, not only ozone peak concentrations need to be examined, but also ozone exposures on "seasonal" scale need to be quantified. Modelling systems can represent suitable tools for this purpose (*Carmichael et al.*, 1986). As a matter of fact, the Italian law designates the integrated use of representative stations, for monitoring activities, emission inventories and modelling systems, for simulating air pollutants transport, transformation and diffusion, as the best approach for air quality assessment. In this study an integrated modelling system has been developed in order to perform a simulation of the photochemical pollution.

The main modules of the modelling system are: the emission pre-processor, the prognostic non-hydrostatic meteorological model RAMS (version 4.3/4.4, **R**egional **A**tmospheric **M**odelling **S**ystem, *Pielke et al.*, 1992), the interface module RAMS-CAMx, which is distributed by Environ as free software (http://www.camx.com/down/support.php) and the Eulerian model CAMx (version 4.20, **C**omprehensive **A**ir Quality **M**odel with e**x**tensions, *Environ*, 2005).

The modelling system has been applied over a domain, in Northwest Italy, covering areas characterized by different emission levels: eastern Piedmont valley (low emission level area) and industrial triangle Milan-Turin-Genoa (high emission level area). This region is often affected by severe summer photochemical pollution episodes driven by both anthropogenic emissions and meteorological conditions. Indeed the Po river valley is very industrialised and populated and it is characterised by strong urban, industrial and traffic emissions; moreover, the presence of the Alps often gives rise to weak circulation and stagnant conditions.

In this paper, the results of the six-summer months simulation (from April to September 1999) are compared with ozone and nitrogen dioxide data measured at several monitoring stations, grouped in urban, suburban and rural sites. The model performances, analysed and discussed in term of statistical indices, show a general satisfactory agreement.

METHODOLOGY

The modelling system includes the meteorological model (RAMS), three interface modules (RAMS-CAMx), an emission processor and the photochemical model (CAMx). RAMS is build up around the full set of the primitive dynamic equations. The model is driven by means of the nudging technique using the ECMWF (European Center for Medium-Range Weather Forecasts) analysis as boundary conditions. These fields are also used to give the initial conditions. Among the vertical diffusivity schemes available, the Mellor and Yamada scheme 2.5 (*Mellor and Yamada*, 1982) has been chosen to compute the vertical mixing coefficients.

The model has been carried out using one single grid. The first interface module provided to CAMx height and pressure for each vertical grid level, horizontal wind components, temperatures, vertical diffusivities, water vapor, clouds and rain. The second module provided topography and land use, and the last one provided the photolysis rates. These rates are derived for each grid cell assuming clear sky conditions as a function of five parameters: solar zenith angle, altitude, total ozone column, surface albedo and atmospheric turbidity.

The emissions processing system is designed by us to produce emission fields according to the model characteristics. The 1999 emissive data sets have been derived from regional emission inventories covering areas within the Italian boundary. EMEP inventory, that provides emissions over Europe on cells 50x50 km^2 has been used for the remaining part of the domain. The necessary processing of the raw data (*Seinfeld*, 1988) has been performed with the following steps: the spatial disaggregation; the temporal disaggregation and the hydrocarbon speciation.

CAMx is an Eulerian photochemical dispersion model that simulates the emission, dispersion, chemical reaction and removal of pollutants in the troposphere by solving the pollutant continuity equation for each chemical species on system of nested three-dimensional grids.

The photochemical model CAMx implements an extended version of the CB-IV mechanism (*Whitten et al.*, 1980, *Gery et al.*, 1989) including aerosol and mercury chemistry and several additional inorganic reactions appropriate for regional modeling conditions. It includes 117 reactions and 67 species.

The model domain (Figure 1) covers an area of 250x250 km^2, including different Northwest Italian regions, a portion of Mediterranean Sea, and some European areas. Therefore a main low-lying plain area (the Po valley) characterizes the domain and the elevated Alpine mountains, the Apennines and the sea surround the domain, in northwest and south direction respectively.

The CAMx simulation area has been horizontally subdivided into 50x50 cells, with a space resolution of 5 km and the vertical domain extends up to 5490 [m agl], subdivided into 14 layers of growing thickness.

The initial and boundary conditions for the model run have been provided by a large-scale simulation (*M.P.Costa et al.*, 2006).

Both meteorological and photochemical models have been run for the whole summer 1999, from April to September supplying hourly concentrations fields. This period was selected because of the severe and critical ozone episodes generally occurring during the summer time. In fact the most high ozone concentrations, that exceed law limit, are detected during the hot months with intense solar radiation.

RESULTS AND DISCUSSION

Ozone and nitrogen dioxide hourly time series from the air quality networks (Figure 1) have been analyzed and, starting from obtained results, a set of eight representative monitoring stations has been selected to compare the results of the simulation with measurements.

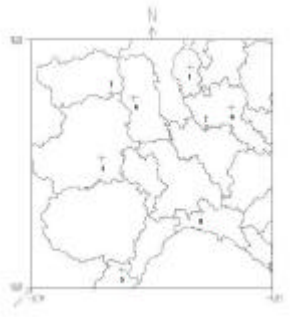

Fig.1; Simulation domain and monitoring.

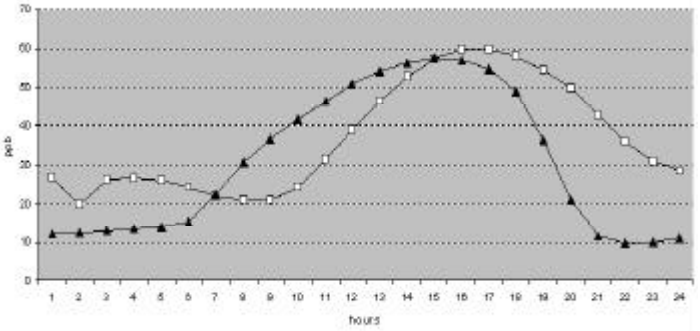

Fig. 2; Mean ozone day at urban sites (st. 3, 4, 6), calculated on the hourly concentrations (□-measured, ▲-CAMx)

The comparison between observed and predicted ozone and nitrogen dioxide levels at different kind of monitoring sites (rural, suburban and urban station) is presented in the following Figures (2, 3 and 4): regarding the ozone and the nitrogen dioxide, the hourly concentration, over the six months, has been taken into account. Then both for O_3 and for NO_2 the stations have been grouped by category (urban, suburban and rural) and then we have calculated the statistical indices (Table 1 and 2): the O_3 and NO_2 indices have been computed on the daily maximum 8-hourly average and the mean daily concentration respectively. The statistical indices used are: the correlation coefficient (R), the average concentration over the six months (MEAN), the normalized mean square error (NMSE) and the fractional bias (FB). For FB, the minus means overestimation and the plus means underestimation.

Figure 2 reports the comparison between measured and simulated mean day as an average for the three urban stations. The figure shows a rather satisfactory agreement. Moreover it can be observed the characteristic diurnal mean ozone pattern, even if the CAMx curve is shifted backward with respect to the measurement one.

In Figure 3, the comparison for six months average ozone concentration is shown: for almost all stations the model underestimates the observed values.

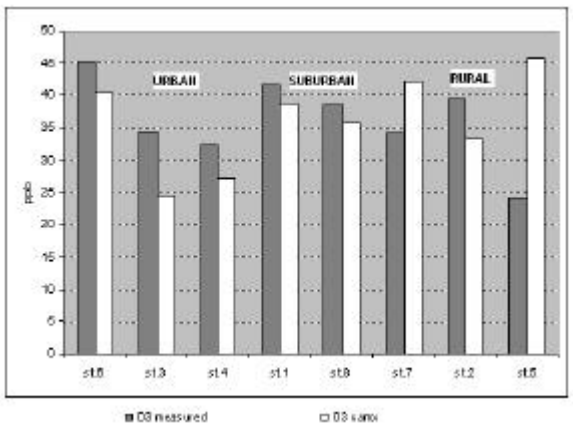

Fig. 3; Mean ozone value at urban (st. 3, 4, 6), suburban (st. 1, 8) and rural (st. 2, 5, 7) sites, calculated on the hourly concentrations.

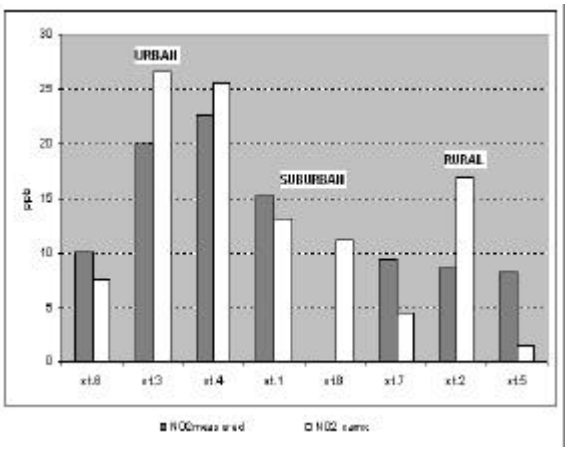

Fig. 4; Mean NO_2 value at urban (st. 3, 4, 6), suburban (st. 1, 8) and rural (st. 2, 5, 7) sites, calculated on the hourly concentrations.

In Figure 4, the comparison for six months average nitrogen dioxide concentration is presented: the model shows both overestimations and underestimations with respect to the

observed values, regardless their category; at site number 8, it was not possible to compare the model results with the measurements because the observed NO_2 data didn't available.

The analysis of the ozone statistical indices (Table 1) demonstrates that, regarding urban and suburban stations, the model underestimates; instead it overestimates at rural sites. It can be observed a rather good correlation between measured and computed values: in particular at suburban station, the simulation has provided a slightly higher correlation coefficient than at the other stations and the computed mean value is closer to the observed one.

Table 1. Ozone indices computed on the maximum 8-houlry average.

station	type	O_3 MEAN (ppb)	R	NMSE	FB
URBAN	meas	58.55	1.0	0.0	0.0
	camx	53.82	0.57	0.06	0.08
SUBURBAN	meas	58.08	1.0	0.0	0.0
	camx	56.72	0.61	0.10	0.02
RURAL	meas	52.91	1.0	0.0	0.0
	camx	56.47	0.59	0.04	-0.07

Table 2. NO_2 indices computed on the mean daily average.

station	type	NO_2 MEAN (ppb)	R	NMSE	FB
URBAN	meas	18.05	1.0	0.0	0.0
	camx	20.34	0.52	0.10	-0.12
SUBURBAN	meas	15.50	1.0	0.0	0.0
	camx	12.12	0.35	0.47	0.25
RURAL	meas	8.95	1.0	0.0	0.0
	camx	7.65	0.24	0.19	0.16

Concerning the nitrogen dioxide (Table 2), the model performances are slightly worse. The urban values measured are overestimated; instead the suburban and rural measurements are underestimated. The correlation coefficient improves when moving from rural to urban stations, even if, in the rural case, the model well reproduces the mean values. As a matter of fact, NO_2 concentrations directly depend on the emissions hence they are more influenced by local scale processes that are likely not well captured at the adopted horizontal resolution.

CONCLUSIONS

A photochemical pollution model system has been applied to simulate airborne dispersion and chemical reactions on a domain with a grid resolution of 5 km. The simulation considers a local scale domain (250x250 km^2) placed in the North-West of Italy. The simulation has been run for six months, from April to September 1999. Simulation results over the six months have been compared with air quality data. Obtained results have shown a general agreement between computed and measured ozone concentrations, even if some discrepancies are highlighted.

The model has also put in evidence some relevant nitrogen dioxide overestimations, mainly in the urban areas, probably due to an underestimation of the vertical diffusivity. As a matter of fact the Mellor and Yamada 2.5 closure implemented in RAMS underestimates the turbulent kinetic energy, as shown by Trini Castelli et al., 2001 and Ferrero et al., 2003. This result could explain also the ozone underestimation. Nevertheless, the model results are considered

encouraging, if the inherent complexity in reconstructing a proper emission picture and in the meteorology of the complex orographic site is taken into account.

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